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ARE 0.1%-ACCURATE GAMMA-RAY ASSAYS POSSIBLE FOR ^{235}U SOLUTIONS?^a

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ABSTRACT

The factors influencing the accuracy of passive gamma-ray assay of uniform, homogeneous solution samples have been studied in some detail, particularly for the assay of ^{235}U in uranium solutions. Factors considered are the overall long-term electronic stability, the information losses caused by the rate-related electronic processes of pulse pileup and deadtime, and the self-attenuation of gamma rays within the samples. Both experimental and computational studies indicate that gamma-ray assay procedures for solution samples of moderate size (from 10 to perhaps a few hundred milliliters) are now capable of accuracies approaching 0.1% in many practical cases.

I. INTRODUCTION

Solutions, because they are uniform and homogeneous, offer the possibility for highly accurate nondestructive gamma-ray assays of the gamma-ray-emitting isotopes contained in them. A task to develop a gamma-ray assay system to determine the ^{235}U content of enriched-uranium solutions has provided the opportunity to study carefully some of the more important factors influencing the accuracy of quantitative gamma-ray assays. The factors studied are the long-term electronic stability of modern assay systems, the information losses resulting from the rate-related electronic processes of pulse pileup and processing circuit deadtime, and the self-attenuation of the emitted gamma rays within the samples. The last factor, sample self-attenuation, is particularly important in solutions of high-Z elements such as uranium because of their very high mass attenuation coefficients.

In discussing the accuracy of gamma-ray assay procedures, it is convenient to also speak of precision and bias. Because various definitions are in use for all three terms, it seems necessary to explain briefly the meaning of the

terms as used in this paper, though with little effort at mathematical rigor. Precision will be essentially pure repeatability, a measure of the dispersion about its average value of a set of assay values from repeated measurements of the same item under specified and constant conditions. Thus defined, a measurement procedure may be highly precise but very inaccurate. The measure of precision will be the standard deviation, the square root of the sample variance [see Eq. (4)]. Bias will mean the deviation of a highly precise average of a set of measurements from the true or accepted value. Accuracy will imply both high precision and low bias. The phrase 0.1% accuracy will essentially mean that 68% of all measurements are within 0.1% of the accepted value and that 95% are within 0.2% of the accepted value.

The design goals for the system motivating the study required the determination of the ^{235}U mass in 30-ml samples with concentrations ranging from 1 to 500 g $^{235}\text{U}/\text{L}$. Throughput requirements dictate a maximum assay time of 1000 s. For solution concentrations >50 g $^{235}\text{U}/\text{L}$, assays require precisions of $\leq 0.1\%$ and accuracies as close to 0.1% as possible. In the lower concentration range, 1-50 g $^{235}\text{U}/\text{L}$, the assay bias is still required to be $<0.1\%$, even though the precision and therefore the accuracy will be $>0.1\%$ for the prescribed assay time. The measurements are based upon the detection of the 185.7-keV gamma ray emitted by ^{235}U at the rate of $\sim 43\,000/\text{s-g}$.

II. FACTORS INFLUENCING ASSAY ACCURACY

The long-term stability (for periods of days at least) of the detector-electronic system used is obviously crucial to obtaining consistently accurate assay results. Based upon past experience, it was immediately concluded that the requested system must employ high-resolution germanium detectors and high-quality electronic data acquisition equipment if 0.1% accuracy is to be approached. The question was whether the stability of such detector-electronic systems is good enough to give overall precisions of $\leq 0.1\%$ for reasonably long time periods. The

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importance of various electronic parameters may vary depending on the specific procedures used for data acquisition and reduction. In the current case involving ^{235}U assay, for example, the high-resolution spectrum is sufficiently uncomplicated to justify the use of simple region-of-interest (ROI) summation methods to extract the full-energy peak areas, which are the fundamental information required. When using fixed ROIs, it is extremely important that the full-energy peaks remain fixed relative to the ROI, which requires that the energy calibration must be very stable, whereas it is not quite as important to have a very stable peak shape as it would be when using some of the spectral fitting codes. For any proposed assay procedure, the important parameters must have the required stability, not just in an environmentally controlled laboratory, but often in production-type environments under quite difficult conditions.

When uranium solutions to be assayed cover a wide range of concentrations as well as a wide range of solution types, the corrections both for the electronic losses caused by deadtime and pileup and for the sample self-attenuation become important. The corrections for the self-attenuation may vary over several hundred per cent, and because count rates generally follow the concentration, though less than proportionally, the corrections for the pileup and deadtime losses may also vary by several hundred per cent. This is particularly true of the pileup and deadtime losses when it is necessary to use short sample-to-detector distances to increase the count rates to obtain good precision in a short time.

As formulated within the nuclear safeguards research and development program at the Los Alamos National Laboratory, quantitative gamma-ray assays are usually based upon a corrected count rate from the isotope of interest given by

$$CR = RR * CF(RATE) * CF(ATT) \quad (1)$$

where CR is the corrected rate, RR is the raw acquisition rate, CF(RATE) is the correction factor for the rate-related electronic losses resulting from the combined effects of pileup and deadtime, and CF(ATT) is the correction factor for the self-attenuation of the gamma radiation within the sample. When the correction factors CF(RATE) and CF(ATT) are properly defined and computed, CR becomes the count rate that would have been observed if there were no electronic losses and no sample self-attenuation. Often, CF(ATT) is defined in such a way that CR is the rate that would have been observed if the sample had been reduced to a non-

attenuating point in a fixed position relative to the detector. With the two correction factors thus defined, CR is proportional to the mass of gamma-ray-emitting isotope, and we have the relationship

$$CR = K * M \quad (2)$$

where M is the mass of the isotope being assayed and K is a constant of proportionality, which is actually the calibration factor for the assay geometry being used. Such linear, one-parameter calibration "curves" are obviously simple to use and are particularly useful when it is desirable to extrapolate somewhat beyond the range of isotope mass represented in the calibration standards. Note also that when CR is defined as above, the calibration is independent of the chemical composition of the standards and unknowns and is quite insensitive to reasonable differences in size and shape between them.

All three factors entering into CR are important. In principle, RR is easiest to determine, being just the full-energy peak area divided by the true time of data acquisition. However, to avoid biased values of the peak area, which are the fundamental data entering into CF(RATE) and CF(ATT) as well as RR, good-quality, modern equipment (including the detector) must be used, and it must be properly set up and adjusted. If this is done, the full-energy peaks should be nearly symmetric and essentially free of either high- or low-energy tailing, which makes it difficult to extract the peak areas in an unbiased manner. The use of pileup rejection improves the peak shape as well as the overall precision and should probably be used when data acquisition is required at high rates.

A. Electronic Stability

Most of the testing of electronic stability was done by examining the pure repeatability of the ratios of full-energy peak areas from appropriate multiple isotope sources. Because the study was motivated by the development of a non-destructive assay system for ^{235}U in solutions, much of the work was done with combined sources of highly enriched uranium metal (~93% ^{235}U) and ^{241}Am , the ^{241}Am being used because it is a strong candidate for use as the reference source for the correction of deadtime and pileup losses in that assay system. When using the reference-source procedure for correction of rate-dependent electronic losses, all results are based on the ratio of full-energy peak areas of the assay isotope and the reference source. Thus, examining the repeatability of such ratios seemed a

legitimate way to test the stability parameters most important to the proposed assay device.

The intent of the exercise was to test only the stability of the electronic portion of the system and to avoid all positional effects, which could occur if sources moved slightly relative to one another or to the detector. Thus, combined sources were used, a ^{241}Am source taped firmly to an enriched-uranium disk in most cases. They were placed far enough from the detectors that a slight change in the position of the combined source relative to the detectors did not significantly change ($\ll 0.1\%$) the ratio of the full-energy interaction rates (FEIRs) for the gamma rays of interest. Only energy-dependent change in the full-energy interaction efficiency of the detector should have been able to change the ratio of FEIRs significantly, and that kind of change is certainly not expected during periods of days or weeks.

So that small instrumental effects could be detected, it was necessary to maximize the data throughput to achieve the best possible statistical precision in the shortest possible time. To do so, gross counting rates of $\sim 35\,000/\text{s}$ were used in all test exercises. The amplifiers were used with 1- μs time constants, and only 1024-channel conversion gains were used in the 100-MHz Wilkinson-type analog-to-digital converters. Under these conditions, even though the spectroscopy systems used for the test were not particularly fast, the full-energy peak areas of both the ^{241}Am 59.5-keV gamma ray and the ^{235}U 185.7-keV gamma ray grew at rates of between 2000 and 3000 events per second depending on the combined source used and its exact position relative to the detector. In a 500-s true-time count, both peak areas had well over 10^6 counts and precisions of $\leq 0.1\%$, the estimated precision of the area ratios was typically $\sim 0.13\%$.

To guarantee that the full-energy peaks would remain fixed in position relative to the ROI used to extract the peak areas, digital stabilization was used in all cases. In all tests involving ^{241}Am and ^{235}U , both the 59.5-keV peak and the 185.7-keV peak were stabilized. With stabilization no peak of interest ever moved more than a few hundredths of a channel from its nominal position, thus effectively eliminating drift in the energy calibration as a matter of concern.

Two totally different (including the germanium detector), though similar, systems were tested. Because of software limitations, the first system was limited to counts of ≤ 600 -s true time to avoid channel overflow. When the

second system was tested, those limitations had been eliminated. Table I summarizes seven sets of data taken with the first system over a period of 13 days using two different combined sources. The computed estimates of the standard deviations of the grand averages are consistent with the values estimated from the counting statistics for data from both sources. Of the seven reduced chi-square values (χ^2/ν), five are greater than 1, and the probabilities for the values found suggest an influence of electronic stability on the overall precision of no more than 0.01%. Table II demonstrates the consistency within one of the 500-count data sets. In this particular set covering 85 h, there is no detectable instrumental influence at even the 0.01% level.

Table III gives results of a test of the second system examined. In this case, with no limitation on count time, 6000-s counts were used giving a precision of $\sim 0.035\%$ for the 185.7- to 59.5-keV area ratios. Sixty spectra were acquired in a continuous 100-h period, and in this case, ratios were also computed for the other ^{235}U gamma rays of 143.8, 164.6, and 203.3 keV. Again the standard deviations computed from the 60 replicate spectra agree very well with those computed from the estimated precisions of the various peak areas. Three of the four χ^2/ν values are >1.00 with probabilities

TABLE I
SUMMARY OF RESULTS OF SEVEN PRECISION RUNS
WITH SYSTEM NO. 1

Number of Spectra in Run	True Time per Spectrum (s)	Average Ratio	Estimated RSD ^a of Ratio (%)	χ^2/ν
125	300	1.2480	0.011	1.132
125	400	1.2477	0.012	0.983
500	380	1.2479	0.005	1.023
125	500	1.2478	0.012	0.985
Calculated RSD			0.011	
125	300	0.8136	0.011	1.001
125	300	0.8138	0.011	1.017
500	600	0.8137	0.005	1.002
Calculated RSD			0.010	

^aRelative standard deviation.

TABLE II
INTERNAL CONSISTENCY OF 1 PRECISION RUN
OF 500 SPECTRA OF 600 s EACH

Calculated RSD of single ratio (%)	0.1162
Av. estimated RSD of single ratio (%)	0.1160
Reduced chi-square	1.004
Av. of 500 ratios	0.81373
Av. of 1st 100 ratios	0.8138
Av. of 2nd 100 ratios	0.8137
Av. of 3rd 100 ratios	0.8138
Av. of 4th 100 ratios	0.8137
Av. of 5th 100 ratios	0.8138

again indicating an instrumental influence of no more than 0.01%.

The results of the exercises reported and of many not described all seem to agree that modern gamma-ray spectroscopy systems are sufficiently stable to give long-term area-ratio precisions of ~0.01%. Such stability and precision is adequate to permit 0.1%-accurate assays if the necessary corrections can also be made with sufficient accuracy.

The χ^2/ν values presented in Tables I, II, and III are computed from the definition

$$\chi^2/\nu = S^2/\sigma^2 \quad (3)$$

The quantity S^2 is the sample variance of the measured peak area ratios from the replicate spectra. It is computed from the formula

$$S^2 = \left[\sum_{i=1}^n (R_i - \bar{R})^2 \right] / (n - 1) \quad (4)$$

where R_i are the measured peak area ratios, \bar{R} is the average ratio, and n is the number of ratios in the data set. The quantity σ^2 is the average estimated variance of the individual R_i , which are based on the estimated variances of the two peak areas concerned. They in turn are computed from the variances of the ROI sums from which the area is found, and the variances of the ROI sums are, under the assumption of Poisson statistics, equal to the sums themselves. Because of the crucial importance to the precision test of properly estimating the variance of the peak area ratios, the relationship used is given here, though without derivation, along with the algorithm used in computing the peak areas. Figure 1 shows schematically a portion of spectrum containing one full-energy peak with the ROI used to find the peak area and its estimated variance along with notation for the required ROI parameters. With the notation from Fig. 1, the background subtracted from the peak ROI sum is

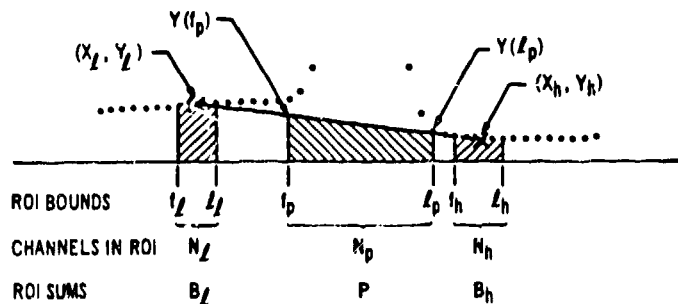
$$B = \left[Y(f_p) + Y(i_p) \right] * \frac{N_p}{2} \quad (5)$$

which is the area in the peak ROI beneath the line defined by the centers of the background ROI and the average count in them. The full-energy peak area is then

$$A = P - B \quad (6)$$

TABLE III
SUMMARY OF RESULTS OF PRECISION TEST OF SYSTEM NO. 2
(60 SPECTRA OF 6000 s EACH)

Gamma-Ray Energy (keV)	Mean Area Ratio to 59.5-keV Gamma Ray	Calculated RSD of Single Ratio (%)	Estimated RSD of Single Ratio (%)	χ^2/ν	Probability for Exceeding χ^2/ν (%)
143.8	0.165	0.087	0.086	1.023	43
164.6	0.101	0.113	0.116	0.957	57
185.7	1.430	0.038	0.036	1.138	22
205.3	0.146	0.079	0.079	1.014	45



where

X_L , X_h are, respectively, the centers of low- and high-energy background ROIs; Y_L , Y_h are, respectively, the average count/channel in the low- and high-energy background ROIs; $Y(f_p)$, $Y(f_p)$ are the ordinates of the background line at f_p and f_p , respectively; and $N_L = f_L - f_L + 1$, $N_p = f_p - f_p + 1$, $N_h = f_h - f_h + 1$.

Fig. 1. ROI and associated parameters used in computing the net background-subtracted area of a full-energy peak and the estimated standard deviation of the area.

Now the expression for the estimated variance of the area is

$$\sigma^2(A) = \sigma^2(P) + \sigma^2(B) \quad (7)$$

where

$$\sigma^2(P) = P \quad (8)$$

and

$$\sigma^2(B) = \left(\frac{N_p}{2}\right)^2 \left[K^2 + \frac{B_h}{N_h^2} + (2 - K)^2 + \frac{B_L}{N_L^2} \right] \quad (9)$$

with

$$K = (f_p + f_p - 2X_L)/(X_h - X_L) \quad (10)$$

The expression for $\sigma^2(B)$, though somewhat complex, is correct for the cases where the background ROIs are not symmetrically placed relative to the peak ROIs. If the background ROIs are symmetrically placed, that is, with

$(f_p - X_L) = (X_h - f_p)$, the coefficient $K = 1.000$, and a simpler expression is obtained. Finally, using standard procedures for combining the variances of a quotient, we have for the estimate of the variance of a peak area ratio, A_1/A_2 , the expression

$$\sigma^2\left(\frac{A_1}{A_2}\right) = \left(\frac{A_1}{A_2}\right)^2 \left[\frac{\sigma^2(A_1)}{A_1^2} + \frac{\sigma^2(A_2)}{A_2^2} \right] \quad (11)$$

The only condition that must apply is that none of the six ROIs involved may overlap, which would destroy the assumed independence of the estimated variances of the ROI sums.

B. Electronic Losses

In high-resolution gamma-ray spectroscopy the loss of information resulting from the combined effects of electronic pileup and deadtime is great at high input count rates, often >50%. Even at low rates, the loss is significant, and corrections must be carefully made at all rates if accuracy approaching 0.1% is to be achieved

in passive gamma-ray assays. There are currently three types of methods for making the necessary corrections. All the methods make the basic assumption that all spectral full-energy peaks suffer the same fraction of loss because of deadtime and pileup.

First, there are the purely electronic methods, most of them based upon fast-timing circuitry that detects piled-up events and extends the counting time sufficiently to compensate for the combined effects of both pileup and deadtime. Such systems suffer limitations in the finite resolving time of germanium detectors (usually >0.5 ns) and in the first-order procedure normally used in extending the counting time. In general, they are not adequate for use in assay systems where accuracy approaching 0.1% is desired over a fairly broad range of count rate. Recent work, particularly by Westphal,¹ seems to remove the limitations and may offer the required accuracy at the price of a more complex data acquisition system. Additionally, if work must be done under conditions of both changing rate and spectral shape, methods such as those of Westphal are the only ones with the required capability.

A second commonly used procedure employs pulsers to inject into the spectrum a peak that suffers nearly the same fraction of losses as do the gamma-ray peaks. Correction is made by comparing the number of pulses injected with the number appearing in the pulser peak. There are numerous implementations of the pulser method, some employing ordinary fixed-period pulsers, some using random pulsers, and some using pulsers in which the pulser rate is related to the input rate to the spectroscopy system. Good results can be obtained with all the implementations, but there are common difficulties relating to amplitude and/or rate stabilities of the pulsers and to the difficulty of inserting the pulses through the preamplifier without some degree of spectrum-distorting undershoot at the amplifier output. If a fixed-period pulser is used, as is most common, a correction must also be made for the fact that the pulser events, which neither pile up on themselves nor are lost because of their own deadtime, suffer somewhat smaller losses than the full-energy gamma-ray events.

The third method may be termed the reference-source method, and at this time (late 1983) it seems most able to provide, with standard spectroscopy equipment, the accuracy needed in the assay system being developed at Los Alamos. It is similar to the pulser method in that corrections are based on a reference peak in the spectrum, but the reference peak comes from a

gamma-ray source fixed in position relative to the detector so that the detector sees a constant flux from the source. Stated differently, the reference gamma ray has a fixed FEIR in the detector. Based on the previously stated assumption that all the full-energy peaks in a spectrum suffer the same fraction of loss because of deadtime and pileup, the ratios of the areas of any other assay-related peaks to the reference peak area are independent of such losses. Assays could be based on such ratios without ever explicitly determining the actual fraction of loss. However, one would usually like to know the actual magnitudes of the rates and corrections involved in an assay, and that desire led to the formulation of the corrected rate as given in Eq. (1).

Because of the nature of the correction test exercises described below, it is worth discussing the form used for CF(RATE) and the form of Eq. (1) resulting from its use. Letting the argument Y refer to any gamma ray other than the reference and the argument R refer to the reference gamma ray only, we can define the correction factor CF(RATE) as

$$CF(RATE) = \frac{FEIR(R) * TT}{A(R)} \quad (12)$$

where

TT = the true time of data acquisition.

Thus defined, CF(RATE) is the ratio of the total number of reference gamma-ray full-energy interactions to the number of them stored in the full-energy peak and is therefore the reciprocal of the fraction stored. The product $RR = CF(RATE)$ in Eq. (1) is then the FEIR of the gamma ray of interest. Letting $RR(Y) = A(Y)/TT$, we have the expression

$$FEIR(Y) = \frac{A(Y)}{A(R)} * FEIR(R) \quad (13)$$

so Eq. (1) can be rewritten in its simplest form as

$$CR(Y) = \frac{A(Y)}{A(R)} * FEIR(R) * CF(ATT) \quad (14)$$

It is seen explicitly in this form that the corrected rates do not depend on the true time of acquisition (though the precision obviously does), a virtue when a fixed assay time is desirable, as it usually is for a high throughput of routine assays. It should be mentioned that the accuracy of assay does not depend on an accurate value of FEIR(R). Indeed, FEIR(R) cancels out when calibration is performed, but a good value enables one to know the actual rates

involved in the assay, which is important when monitoring the performance of a system.

The other strengths and limitations of the reference-source method should be mentioned. Certainly its simplicity is a virtue in that the procedure can be applied to any spectroscopy system without the addition of more electronics. There are no problems in injecting pulser outputs into the preamplifier, there are no corrections to the corrections as there are in using a fixed-period pulser, and there is no difficulty because of the finite pulse-pair resolution time of the timing circuits. In addition, the reference peak is constantly present for digital stabilization and for the checking of detector performance. A significant limitation is that finding a source with appropriate half-life and gamma-ray energy for a desired application is not always possible. In addition, the reference source itself must usually have a significant count rate that causes additional losses and results in poorer overall precision than would be achieved in the same counting time with other methods. And finally, the reference-source method (as well as the simpler pulser procedures) is only applicable in situations of constant count rate and constant spectral shape.

The assumption that all the full-energy peaks suffer the same fraction of loss is not exactly true in practice, primarily because the width and detailed shape of the peaks are functions of both energy and count rate. In applying the reference-peak method to correct for rate-dependent losses, precautions must be observed to minimize the degree to which the assumption falls short. Some of those precautions, most of which would apply to any of the correction methods, are listed below.

- (1) Apply the procedure, where possible, only over a narrow energy range.
- (2) Try to keep the peak width and shape as constant as possible as functions of both energy and count rate, even if that requires degrading the low-rate peak resolution to some extent. Proper adjustment of the amplifier and the use of pileup rejection can help considerably.
- (3) Avoid convex or concave backgrounds beneath important peaks, especially the reference peak. It is good if the ratio of the reference peak area to the background area beneath it can be kept ≥ 10 .

- (4) Great care must be exercised in extracting the full-energy peak areas. No lengthy discussion can be given, but obtaining accurate peak areas unbiased by variation in peak width or minor rate-dependent tailing is crucial to obtaining accurate values of $CF(RATE)$ and to overall accuracy. It appears that ROI methods for computing the areas may be less sensitive to small changes in peak shape than are some of the spectral fitting codes when the spectra are sufficiently simple to use the ROI schemes.

The results of two tests of the accuracy with which pileup and deadtime can be corrected will be presented here. The first test (done in 1976) used solution samples of ^{75}Se of identical attenuation and shape but with different total ^{75}Se activities. The absolute activities were only approximately known, but the relative activities were known to $\leq 0.2\%$. The 25-ml samples were in bottles similar to those to be used in the ^{235}U assay system now under development and were counted at a sample-to-detector distance of ~ 10 cm, great enough to make negligible any variations in count rates resulting from variation in bottle dimensions. The reference source was ^{109}Cd , which has a single gamma ray with an energy of 88.0 keV. Standard high-quality spectroscopy equipment was used including pileup rejection and digital stabilization. The reference source-sample combinations gave gross counting rates over a range of ~ 9000 s^{-1} to $\sim 60\,000$ s^{-1} . The full-energy peak areas were computed by ROI methods using straight-line background subtraction. Figure 2 gives the test results for the 136.0-keV gamma ray of ^{75}Se , the one that had the greatest intensity and thus the best precision. The lower portion of the figure shows the magnitude of $CF(RATE)$ as a function of gross count rate, and $CF(RATE)$ is seen to change from ~ 1.1 to ~ 4.4 , a change of $\sim 300\%$. The upper portion of the figure gives the fractional deviation from the average of the 136.0-keV PEIR per unit activity, which should have been constant if the corrections were bias free. Although a slight energy-dependent trend is evident, the variations are within $\pm 0.6\%$, which is good considering that the corrections varied by $\sim 300\%$. In this case, the slight trend is due to a rate-dependent, slightly concave background under the 88.0-keV reference peak, a result of nearby higher energy peaks from the ^{75}Se . The increasing concave background caused an increasing negative bias in the full-energy peak area computed for the 88.0-keV peak by the straight-line background method, which in turn caused the increasing error in the ratio of the 136.0- to 88.0-keV areas evident in Fig. 2. Greater low-energy filtering of the

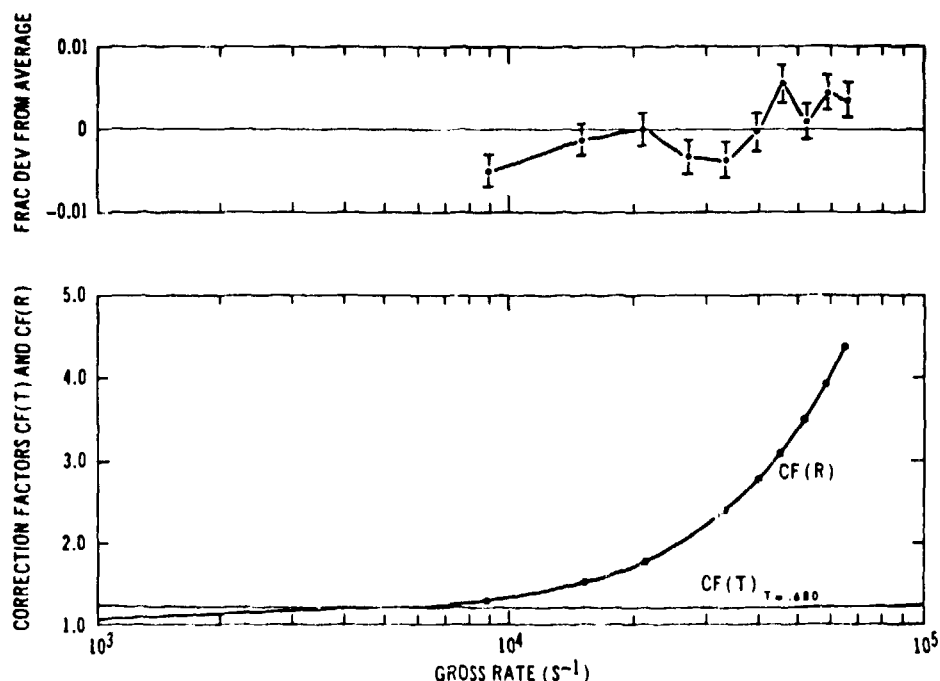


Fig. 2. Results of a test for the accuracy of the correction for counting losses caused by electronic pileup and deadtime. CF(R) is the correction being tested. CF(T) is the constant correction for gamma-ray self-attenuation.

^{75}Se gamma rays and a more intense reference source could have effectively eliminated that problem.

The second test was recently performed and largely avoided the minor problems evident in the first exercise. Seven nominally identical nickel-plated uranium metal disks ($\sim 93\% \text{ }^{235}\text{U}$) were used as counting samples, and ^{241}Am was used as the reference source. A fixture allowed precisely reproducible positioning of each of the seven uranium samples with respect to the detector, and the reference source was, as usual, taped firmly to the detector end cap. Standard high-quality spectroscopy equipment was again used including pileup rejection and digital stabilization. The ^{241}Am reference source alone gave a gross rate of $\sim 9500 \text{ s}^{-1}$ with a 59.5-keV FEIR of 5987 s^{-1} . A single uranium disk added $\sim 6500 \text{ s}^{-1}$ to the gross rate with a 185.7-keV FEIR of $\sim 2300 \text{ s}^{-1}$. With all seven disks in place, the maximum gross rate was $\sim 60\,000 \text{ s}^{-1}$ with a 185.7-keV FEIR of $\sim 16\,000 \text{ s}^{-1}$.

All the background ROIs were three channels (0.6 keV) wide. Because there was a little pileup-induced high-energy tailing of the 185.7 keV peak at the higher rates, the high-energy background ROI was centered at 191.5-keV, beyond the pileup tail. The 185.7-keV low-energy background ROI was centered at 181.1 keV on the low-energy side of the weak but interfering ^{235}U gamma ray at 182.7 keV, and the peak ROI was centered on the peak and was three times wider than the maximum FWHM (19 channels, or 3.8 keV). The 59.5-keV reference peak had negligible high-energy tailing, and the background ROIs were placed symmetrically with 1-channel gaps between them and the peak ROI, which had the same 19-channel width as the 185.7-keV peak ROI. A simple, smoothed-step background subtraction was used in computing the peak areas because it is more accurate when background ROIs must be relatively far from the peaks concerned.

Each disk was counted by itself and the 185.7-keV FEIRs were determined with estimated relative uncertainties of $\pm 0.05\%$.

again determined with relative precisions of $\sim 0.05\%$. Because of the counting fixture, each disk was in precisely the same position relative to the detectors whether counted singly or as part of a combination and therefore produced the same FEIR in the detectors in both cases. The essence of the test was to see whether the measured 185.7-keV FEIRs of the various combinations were equal to the corresponding sums of the 185.7-keV FEIRs of the disks measured singly.

Table IV summarizes the results of the test. The total correction factors, CF(RATE), range from 1.35 to 2.68, thus varying by about a factor of 2. The ratios of the FEIRs of the combinations to the corresponding sums of singly counted FEIRs, which are determined with relative precisions of $\sim 0.06\%$, differ from 1.000 by ≤ 0.001 . It appears that at the highest rates there is an $\sim 0.1\%$ decrease in the ratios. A similar exercise was performed with the uranium samples farther from the detector so that the maximum gross counting rates were $\sim 38\,000\text{ s}^{-1}$ with results that exhibited no discernible systematic trend.

The results given indicate that with standard high-quality spectroscopy equipment, properly set up, and with due care in extracting the full-energy peak areas, the reference-source method can make corrections for the combined losses caused by pileup and deadtime with accuracies approaching 0.1% over a wide range of count rate.

C. Self Attenuation Losses

If the sample linear attenuation coefficient μ^L is known along with the sample dimensions, the container μ^L , the container dimensions, and the position of the container with respect to the detector, the correction for the absorption by the sample of its own radiation, CF(ATT), can usually be computed with adequate accuracy. If the sample-to-detector distance is comparable to or less than the dimensions of the detector or sample, CF(ATT) can usually be computed with less error if the full-energy interaction efficiency of the detector is known as a function of energy and source position relative to the detector. The CF(ATT) are in general strong functions of the sample μ^L and usually much milder functions of the dimensional and positional parameters. However all the parameters must be known with sufficient accuracy to guarantee the necessary accuracy to the computed values of CF(ATT).

1. Error in the Value of Sample μ^L .

If the composition of solution samples is variable, especially with highly variable concentrations of high-Z components, the best, and usually the only accurate way, to determine the sample μ^L is by a measurement of the gamma-ray transmission through the sample. The fundamental law of gamma-ray attenuation is

$$I = I_0 e^{-\mu^L x} \quad (15)$$

TABLE IV

RESULTS OF TEST OF REFERENCE SOURCE PROCEDURE FOR CORRECTION OF COUNTING LOSSES CAUSED BY DEADTIME AND PILEUP

Samples	Approximate Gross Rate (s^{-1})	186-keV FEIR (s^{-1})	Total Correction Factor	Ratio FEIR Σ / Σ FEIRs	σ_r (Ratio) (%)
1-7	60 000	16 353	2.68	0.9990	0.045
1-6	53 200	14 104	2.43	0.9997	0.061
1-5	46 300	11 847	2.19	1.0000	0.061
1-4	39 900	9 703	1.98	0.9997	0.061
1-3	33 700	7 664	1.80	0.9997	0.061
1-2	27 300	5 623	1.62	1.0004	0.067
2 only ^a	16 500	2 135	1.35	--	--

^aRates and correction factor for source No. 2 alone are included only to facilitate comparisons of ranges of rates and correction factors.

where I_0 is the intensity of a beam of gamma rays incident on a slab of material of linear attenuation coefficient μ^L and thickness x , and where I is the intensity of the beam emerging unscattered and unabsorbed. The transmission is defined as the ratio

$$T = I/I_0 = e^{-\mu^L x} \quad (16)$$

The transmission is measured by determining the FEIR of the chosen gamma ray from an appropriate external source I_0 , then by measuring the FEIR again with the sample of known thickness in the gamma-ray beam I . The measured transmission I/I_0 is equated to $\exp(-\mu^L x)$ and solved for μ^L , yielding

$$\mu^L = \frac{-\ln T}{x} \quad (17)$$

The experimentally measured value of μ^L is then used in computing CF(ATT). Because of the generally logarithmic nature of the forms for CF(ATT) as a function of T , the fractional error

in CF(ATT) is always less than one-half the fractional error in the value of T used to derive μ^L , a situation easing somewhat the requirements for accuracy in determining T .²

Figure 3 presents the results of a set of test measurements (performed in 1976) designed to explore the possibilities of accurately computing CF(ATT) over a wide range of measured transmission. In this test, as in that whose results are given in Fig. 2, the samples were 2.5-cm-thick (25 ml in flat-bottomed cylindrical bottles of 10 cm² area) solutions of uranyl nitrate spiked with ⁷⁵Se. The ⁷⁵Se activity was the same to better than 0.1% in all the samples, but the uranium concentration varied from ~0.5-g/l to ~500 g/l. Because of the constant activity, the range of CF(RATE) varied only ~10% over the whole range of samples, whereas the CF(ATT) varied by ~300%, the exercise thus being essentially a test of the accuracy of CF(ATT). The sample-to-detector distance of ~10 cm was sufficient to reduce any error caused by expected variation in the bottle dimensions to <0.1%.

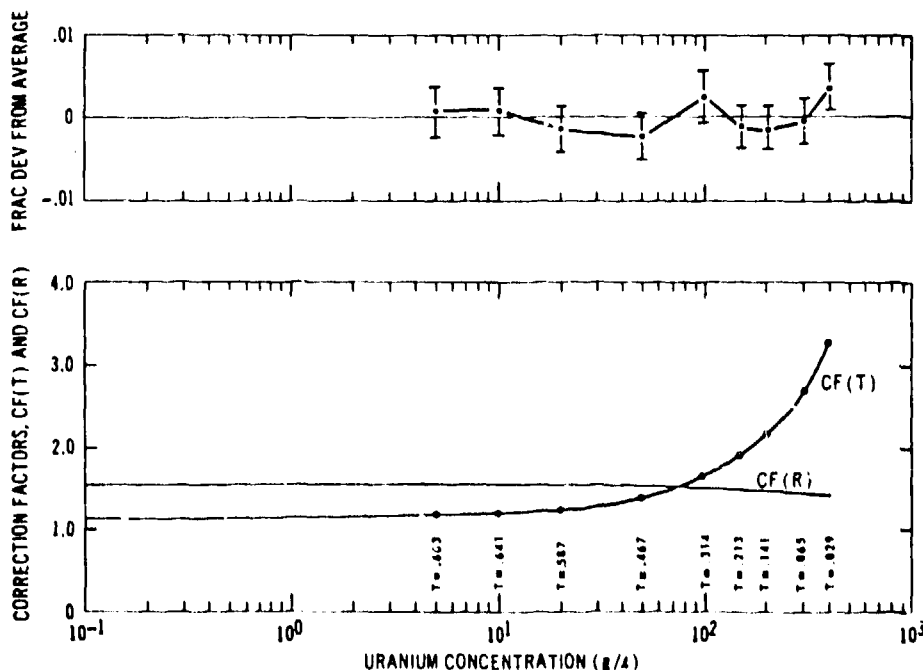


Fig. 3. Results of a test for the accuracy of the correction for information loss caused by gamma ray self-attenuation within the assay samples. CF(T) is the correction being tested. CF(R) is the nearly constant correction for deadtime-pileup losses.

The CF(ATT) were computed by numeric integration on the basis of a simple model using the measured values of U^L . The results presented are for the 136.0-keV gamma ray of ^{75}Se , which had the best precision among the five gamma rays measured as well as high values of CF(ATT).

The lower part of Fig. 3 gives the magnitudes of both CF(RATE), labeled just CF(R) in the figure, and CF(ATT), labeled just CF(T) in the figure, along with the measured values of T. The upper part gives the fractional deviations from the average corrected rate with error bars indicating the estimated one-sigma uncertainty of the points. The deviations are all within 0.5% of the average with no evidence of systematic trend. In spite of the relatively inadequate 0.25% uncertainty of the points, the results show that the sample U^L can be determined by the transmission method with sufficient accuracy to calculate CF(ATT) values with accuracies no worse than a few tenths of a per cent.

2. Error in Dimensions and Position.

In computing CF(ATT) the best average values available for the container dimensions, sample depth, and sample position are usually used because careful individual measurements of every sample would generally be prohibitively expensive. The expected normal variation in dimensions in the manufacturing process will then be a source of error, perhaps random, perhaps systematic, depending on the nature of the variations from the assumed values. In seeking ac-

curate assays, the magnitude of error possible because of such variations must be understood. Because of the difficulty in obtaining containers with slightly different but accurately known dimensions and because of the very high-precision gamma-ray spectral measurements that would be required to measure the effects of the small dimensional differences, a computational approach seemed best to obtain the required understanding.

Because the sample-to-detector distance to be used in the uranium solution system being developed must be quite small to obtain high-precision assays in a relatively short time, it was appropriate to include a model of the detector efficiency as a function of both energy and source position. The procedure described by Cline³ was used to characterize a detector similar to those to be used. Several different sample configurations were modeled using numeric integration for computation. The primary parameters considered in each configuration were uranium concentration, sample radius, sample depth, and sample-to-detector distance, the last parameter obviously being subject to variation because of differences in the thickness of the bottoms of the sample containers.

Table V summarizes results for one sample configuration and concentration of the many combinations studied. It is for a solution 20-cm^2 in area and 1.5 cm deep (30 ml) at a concentration of 100 g U/l. The estimated assay errors in per cent for 0.025-mm (0.001-in.) errors in

TABLE V

ESTIMATED PRECISION^a AND ESTIMATED ERROR FOR A 0.025-mm ERROR
IN GEOMETRICAL PARAMETERS FOR A 30-ml SAMPLE^b
of 100-g/l ^{235}U SOLUTION

Sample-to-Detector Distance (cm)	Precision (%)	Estimated Assay Error for 0.025-mm Errors		
		Sample Depth (%)	Vert. Position (%)	Sample Radius (%)
10	0.17	-0.017	-0.038	-0.0057
5	0.12	-0.026	-0.061	-0.014
3	0.10	-0.033	-0.080	-0.024
2	0.096	-0.037	-0.093	-0.037
1	0.073	-0.042	-0.111	-0.046
0.1	0.005	-0.048	-0.133	-0.055

^aAt one-sigma level in 1000 s.

^b $20\text{ cm}^2 \times 1.5\text{ cm}$ deep.

sample depth, vertical position (sample-to-detector distance), and sample radius are tabulated for various distances up to 10 cm. The estimated precision for 1000-s assays of solutions made from the usual highly enriched uranium available at Los Alamos are also given. It is seen that for distances of 2-3 cm precisions of $<0.1\%$ are possible with positional errors of $<0.1\%$ if the vertical position is controlled to <0.025 mm.

The important point from Table V is that dimensional parameters must be closely controlled if accurate assays are to be obtained at small sample-to-detector distances. It is also clear that if greater counting times can be used to obtain the desired precision, counting with greater sample-to-detector distances is advantageous because of the smaller effects of error in all of the position parameters.

D. Combined Effects of Self-Attenuation and Rate-Related Losses

The major corrections made for each assay are those for the sample self-attenuation, CF(ATT), and the rate-related electronic losses resulting from pileup and deadtime CF(RATE). Generally, no explicit correction for error caused by variation in sample dimension and position will be made, but the magnitude of error or bias related to those effects will be held below acceptable limits by strict control of the pertinent dimension and position parameters. The correction factors CF(ATT) and CF(RATE) are nearly independent, there generally being only mild coupling because of common reference peak areas used in determining them. In actual assay situations, both may vary over wide ranges and both must be correctly determined to get accurate assay results. The development of the uranium solution assay system has not yet reached the point where tests with enriched-uranium solutions for the accuracy of the product $CF(ATT) \times CF(RATE)$ have been performed with the

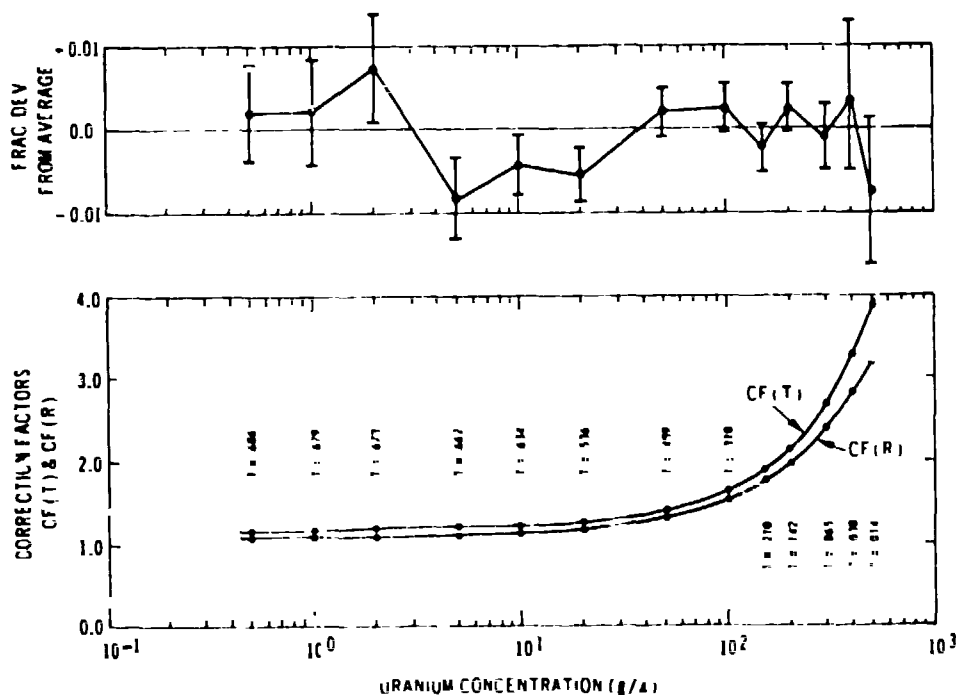


Fig. 4. Results of a test for the accuracy of simultaneous corrections for data losses caused by deadtime-pileup and sample self-attenuation. CF(T) indicates the self-attenuation correction and CF(R) indicates the deadtime-pileup correction. Measured values of the 136.0-keV transmission through the 13 samples are indicated. The upper portion of the figure gives the fractional deviations of the total corrected count rates per unit ^{75}Se activity from the average.

best currently available equipment and procedures. The results of such a test done in 1976 for a similar application show, however, that even then the error of the combined corrections could be kept within a few tenths of a per cent. It is hoped that with improved equipment and procedures the capability can be improved somewhat. Figure 4 summarizes the results of the 1976 test exercise.

As with the previously discussed exercise for the accuracy of CF(ATT), the samples were 25-ml solutions (10 cm² x 2.5 cm deep) of uranyl nitrate spiked with ⁷⁵Se, but in this case the ⁷⁵Se spike was proportional to the uranium concentration, with the amount adjusted to give count rates typical of low-burnup plutonium solutions of the same concentration. The concentrations in the 13 samples ranged from 0.5 to 500-g U/kg, and the ⁷⁵Se activities were known with a relative accuracy of ~0.2%. Again, the results are reported for the 136.0-keV gamma ray of ⁷⁵Se.

The lower portion of Fig. 4 gives the magnitudes of both correction factors as a function of uranium concentration. The measured transmission through the samples varied from ~69% down to ~1.5%, giving rise to values of CF(ATT) between ~1.2 and ~3.8. The gross count rates for the samples alone ranged up to ~70 000 s⁻¹ for the highest concentration, resulting in values of CF(RATE) between ~1.1 and ~3.3. Over the range of concentration and count rate, the product CF(ATT) * CF(RATE) changes by a full factor of 10. The upper portion of the figure gives the fractional deviation of the total corrected count rate per unit ⁷⁵Se activity from the average, with one-sigma estimates of the precision of each point indicated by the error bars. All the deviations are within ±1% of the average.

Considering the uncertainty in the relative ⁷⁵Se activities and the larger-than-desirable precisions of the deviations, the results are good when compared with the ~1000% change in the total correction factor.

This and all other experimental tests and computational studies suggest that for solution samples, with the advantages of uniformity and homogeneity that they bring, gamma-ray assay procedures can have accuracies in the vicinity of 0.1%. Whether the practical limit will be one-tenth of a per cent or several tenths of a per cent only time and more development work will tell. It is hoped that the required work will be largely done during the next year.

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